960,448

PATENT SPECIFICATION

NO DRAWINGS

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960,448

Date of filing Complete Specification May 14, 1962.

Application Date Feb. 27, 1961. *No. 7031/61.*

Complete Specification Published June 10, 1964.

Index at acceptance: —C4 S(3, 4B, 4D2, 4Q1, 4QY, 4V)

International Classification:—C 09 k

COMPLETE SPECIFICATION

Improvements in or relating to Scintillators

We, UNITED KINGDOM ATOMIC ENERGY AUTHORITY, London, a British Authority, do hereby declare the invention, for which we pray that a patent may be granted to us, and the method by which it is to be performed, to be particularly described in and by the following statement:—

This invention relates to scintillation detectors in which naphthalene derivatives are used as a base, covering a range of refractive indices overlapping the lead optical glasses, and to a detector which comprises a mixture of glass particles with a liquid scintillator, the liquid scintillator having a refractive index which matches that of the glass.

For detecting and measuring nuclear particles, liquid scintillators can be used. These scintillators usually comprise a solvent containing a primary solute or "activator" and a secondary solute or "spectrum shifter". The activator serves to collect the energy absorbed by the solvent, and the spectrum shifter converts the ultraviolet light emitted by the activator into visible light which is detectable by a photomultiplier tube. Examples of commonly used solvents are xylene and toluene. A common activator is PPO (2, 5 diphenyloxazole), and a common spectrum shifter is POPOP (1, 4 - bis 2 - (5 - phenyloxazolyl) - benzene).

When it is desirable to detect high-energy gamma rays with a liquid scintillator detector, such as the common xylene or toluene based detector, the dimensions and density of the detector must be sufficient to absorb the energy of the incident gamma rays. With high-energy gamma rays, scintillating crystals of the required size cannot be obtained, but liquid scintillators can be used. However, the density and atomic number of these above prior liquid scintillators are relatively low, necessitating the use of very large volumes. This in turn means that the output due to background radiation may be prohibitively large. However, the density of 1-methyl naphthalene of the liquid detector to be described below is 19% greater than that of toluene. For large scintillation detectors, its use can reduce the volume required by a factor of 1/1.6. Thus, the inherent problem of background radiation in conventional detectors becomes less of a problem in the 1-methyl naphthalene based liquid scintillator of the present invention.

The volume requirement can be further reduced by the use of lead glass particles which are mixed with the 1-methyl naphthalene based liquid scintillator having a refractive index which matches that of the glass.

The gamma ray detection efficiency of conventional liquid detectors at several Mev energy is relatively low and limited because of the lower pair production cross section due to the lower stopping power of the detector at these energies. However, the combined glass particles and liquid detector of this invention, to be described below, is very efficient in detecting high-energy gamma rays in the 5—10 Mev range, for example, because the stopping power of the detector, with its corresponding greater pair production cross section, increases with increasingly greater gamma ray energies in the above range rather than falling as in the organic scintillators.

According to the present invention, a liquid scintillator comprises a liquid alkyl naphthalene solvent containing primary and secondary solutes.

The solvent may be 1-methyl naphthalene, the primary solute PPO (2, 5 diphenyl - oxazole), and the secondary solute POPOP (1, 4 - bis 2 - (5 phenyloxazolyl) - benzene). The PPO concentration may be in range 5—40 gm/litre and the POPOP concentration 0.1—1.0 gm/litre, the preferred concentration

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being about 10 gm/litre and about 0.1 gm/litre respectively.

Also according to the present invention a scintillator comprises a liquid scintillator as aforesaid additionally comprising glass particles mixed with the liquid scintillator, the glass and the liquid scintillator having matched refractive indices.

With 1-methyl naphthalene as the solvent, the glass particles may be of medium flint glass, the liquid containing a small amount of an alkyl benzene to match the refractive index of the liquid scintillator to that of the glass. The medium flint glass may be approximately 42% PbO by weight with a density of about 3.6 and a refractive index of about 1.617, the alkyl benzene being toluene.

The liquid/glass volume ratio may be about 40/60, and the size of the glass particles may be in the range 0.003 to 0.008 inch. Means may be provided for controlling the temperature of the liquid.

In the liquid scintillator of this invention, the preferred solvent is 1-methyl naphthalene and the preferred primary and secondary solutes are PPO and POPOP, respectively. The scintillating liquid may be contained in a conventional spherical housing and is usable in sizes up to about 50 inches mean diameter without excessive background rates. The size of the liquid scintillator may range from about 2 inches mean diameter to the 50 inch size, depending on the use for the scintillator.

The concentration of the primary solute PPO may vary from 5 to 50 gm/litre and the range 5—30 gm/litre will give pulse heights within 5% of the maximum. The mean dispersion ($n_C - n_F$) of a typical 1-methyl naphthalene scintillator solution preparation varied from 0.0165 to 0.0183 as a function of the PPO concentration. In purer samples of this scintillator solution, the mean dispersion can be as much as 0.028. The concentration of the secondary solute POPOP may vary from 0.1 to 1.0 gm/litre. The concentration ranges set forth above for the solutes are not critical and are given by way of example only. In some cases it may even be desirable to exceed them for reasons such as adjustment of the optical dispersion. It has been determined, however, that values of 10 gm/litre PPO and 0.1 gm/litre POPOP in 1-methyl naphthalene will produce near optimum scintillation intensity.

It has also been determined that a pulse height increase of about 10% can be achieved using no PPO primary solute but 0.5—1.0 gm/litre POPOP as the only solute.

The liquid scintillator described above is not limited to PPO and POPOP as the solutes since most other common solutes could be used (although it is known that terphenyl is not satisfactory as the primary solute). Also, 1-ethyl naphthalene can be used as the solvent in place of the 1-methyl napthalene.

The refractive index D of 1-methyl naphthalene scintillator solutions is about 1.618. Such a scintillator can satisfactorily be used for detecting high-energy gamma rays. As pointed out above, for large scintillation detectors, the use of 1-methyl naphthalene as the solvent will reduce the volume required by a factor of 1/1.6.

Although the 1-methyl napthalene based liquid scintillator is superior to the xylene or toluene based scintillator for use with high-energy gamma rays, it still is not as efficient as desired. It has been determined that a combined mixture of lead glass particles with a 1-methyl naphthalene based liquid scintillator will considerably reduce the volume required while at the same time provide a scintillator that is at least four times more efficient in detecting gamma rays in the Mev range.

It is important that the refractive indices of the glass particles and the liquid scintillator should match, in order that the light from the scintillations should follow a path of minimum length to the surface of the scintillator. If they do not match, the light is refracted at the surface of each glass particle and follows a devious path to the surface, in the course of which much of the light is absorbed.

The refractive indices of the flint glasses range from about 1.53 to over 1.9. The density (2.5—6.0 gm/cc) is much higher than liquid scintillator densities, providing considerably greater stopping power for penetrating radiation, particularly gamma rays of several Mev energy. One suitable glass that can be used in the present invention is a medium flint glass which has about 42% PbO by weight, about 48% SiO_2, plus Na and Ca. This glass with the 42% lead content (with a density of about 3.60 and refractive index of 1.617) provides an appreciable pair production cross section above a few Mev of gamma ray energy, so that the stopping power increases between, say, 5 and 10 Mev rather than falling as in the organic scintillators. The improved pair process also improves the probability of producing a full energy scintillation as compared with the Compton process.

The 1-methyl naphthalene based scintillator liquid, with a D index of about 1.618, as discussed above, is used to mix with the glass particles in constructing the combined glass and solvent scintillator of this invention. The refractive index of the 1-methyl naphthalene can be adjusted to that of the glass by dissolving in this solvent a small amount of a liquid of lower refractive index such as toluene which does not interfere with the scintillation process. By using different proportions of 1-methyl naphthalene and an alkyl benzene (such as toluene) it would be possible to match glasses having refractive indices in the range 1.53 to 1.617. However, for use in detecting high-energy gamma rays, the preferred matching index is near the 1.617 value, correspond-

ing to glass densities of about 3.6 and lead oxide content near 42%.

The mean dispersion of the glass should be matched as close as possible with that of the liquid scintillator. For a flint glass of density 3.6, the mean dispersion is given as 0.017. The corresponding value for the liquid scintillator, as discussed above, ranges from 0.0165 to 0.028, giving a mismatch less than 0.012. The mismatch in dispersion for 90° mean deflection at the extremities of the emission band of 1000—4000 angstroms is ± 0.018, so the match attainable is more than adequate.

The size of glass particles used will depend on the particular use of the scintillator. For example, to detect gamma rays of a few hundred kilovolts, a particle size near 0.0003 inch and a small volume detector would be required. For neutron capture work (5—10 Mev energies) the optimum particle size would lie in the range 0.003 to 0.008 inch. As the principle of operation is to sample the electron tracks at random, uniformity of particle size is not necessarily important. However, for purposes of computing the optical properties, it would be desirable to have the particles as uniform as possible. For detecting 5—10 Mev gamma rays, the glass particles should not be made smaller than 0.003 inch for two reasons. First, the 0.030 Mev beta and 0.047 Mev gamma radiation from any RaD (Pb-210) contaminating the lead in the glass, would contribute directly to the background if the particles were much smaller. Secondly, the paths travelled by the scintillation light (and hence its attenuation by absorption) would be increased in length if more interfaces had to be traversed as would be the case if the particles were smaller.

The mixture of the glass and liquid scintillator should contain as much glass and as little liquid as possible, to give the maximum gamma ray absorption. The liquid/glass volume ratio can be about 40/60. The detector can be assembled by adding the liquid scintillator to a vessel full of the glass particles, taking care to exclude bubbles. The overall average density of such a detector will be near 2.4 gm/cc, nearly three times the density of the usual liquid scintillators. The secondary electrons will lose most of their energy in the glass and the light output will be only about one-fifth that of the liquid scintillator alone. However, for gamma rays of several Mev energy, this light output will still provide adequate phototube response.

A glass-liquid spherical detector of about ten-inch diameter (source of gamma rays at the centre) will rival the performance of the large (40—50 inch) all-liquid detectors. A 30-inch glass-liquid spherical detector will provide over 94% total efficiency for detecting gamma rays. For some neutron capture gamma ray detection work, a flight path (hollow tube) through the detector is required. For ease of fabrication and to minimise losses through the tube, an equivalent truncated cylinder could be used instead of a sphere. As an example of such a cylinder, its dimensions could be 80cm × 80cm with a 10cm diameter axial hole. A detector of this size would provide a minimum total efficiency of about 92% for detecting gamma ray energies up to 10 Mev.

In the combined glass-liquid scintillator, some degree of temperature control may be needed to provide and maintain an adequate match of the refractive indices of the glass and liquid because the scintillation liquids show a temperature variation of refractive index several times larger than for the usual glasses.

If the absorption mean free path for the light is large compared to the scintillator dimensions, the temperature variations should not exceed about $\pm 40°$ C around the match point. If the absorption mean free path for the scintillation light is more nearly comparable to the scintillator dimensions, an order of magnitude closer temperature control (i.e. about $\pm 4°$ C) would be needed and can be easily achieved. This temperature control may be achieved by the standard procedures of thermal insulation, a copper liner, and electrical heaters thermostatically operated a few degrees above room temperature, which can readily control to a tenth of a degree or better. As the relative change in refractive index for 1-methyl napthalene is 0.00027 per degree centigrade, it can be seen that very precise temperature control is not required to provide an adequate match of the refractive indices.

The glass used in the above scintillator may contain naturally radioactive materials such as potassium, thorium, and uranium. However, for a usable detector, the background from these materials should be below, say, 1000 c/s. Such a counting rate corresponds to 600 ppm K, 450 ppm U, and 1.5 ppm Th for the main high-energy component for a 200 litre scintillator.

It should be noted that glasses other than flint glasses could be used in the detector. For example, boron-containing or gadolinium-containing glasses could be used for neutron measurements.

WHAT WE CLAIM IS:—

1. A liquid scintillator comprising a liquid alkyl napthalene solvent containing primary and secondary solutes.

2. A scintillator as claimed in claim 1, wherein said solvent is 1-methyl naphthalene, said primary solute is PPO (2, 5 diphenyloxazole), and said secondary solute is POPOP (1, 4 - bis 2 - (5 phenyloxazolyl) - benzene).

3. A scintillator as claimed in claim 2, wherein said PPO has a concentration in the range of 5—40 gm/litre, and said POPOP

has a concentration in the range of 0.1—1.0 gm/litre.

4. A scintillator as claimed in claim 3, wherein said PPO concentration is about 10 gm/litre, and said POPOP concentration is about 0.1 gm/litre.

5. A scintillator as claimed in claim 1 additionally comprising glass particles mixed with the liquid scintillator, the glass and the liquid scintillator having matched refractive indices.

6. A scintillator as claimed in claim 5 wherein the solvent is 1-methyl naphthalene and wherein the glass particles are of medium flint glass, the liquid containing a small amount of an alkyl benzene to match the refractive index of the liquid scintillator to that of the glass.

7. A scintillator as claimed in claim 2 or claim 3 additionally comprising medium flint glass particles mixed with the liquid scintillator, the liquid containing a small amount of an alkyl benzene to match the refractive index of the liquid scintillator to that of the glass.

8. A scintillator as claimed in claim 6 or claim 7 wherein the glass particles are approximately 42% PbO by weight with a density of about 3.6 and a refractive index of about 1.617, and the alkyl benzene is toluene.

9. A scintillator as claimed in claim 8 wherein the liquid/glass volume ratio is about 40/60.

10. A scintillator as claimed in claim 9 wherein the size of the glass particles is in the range 0.003 to 0.008 inch.

11. A scintillator as claimed in any one of claims 5 to 10 comprising means for controlling the temperature of the liquid.

12. A scintillator substantially as hereinbefore described.

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Leamington Spa: Printed for Her Majesty's Stationery Office by the Courier Press.—1964.
Published at The Patent Office, 25, Southampton Buildings, London, W.C.2, from which copies may be obtained.